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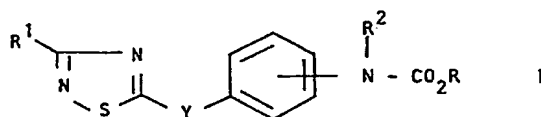
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(54) **Acaricidal 1,2,4-thiadiazoles and their preparation.**

(57) The invention provides the acaricidal thiadiazoles of the formula:



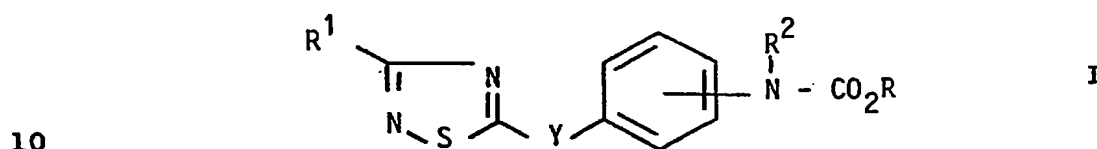
(where: R¹ and R², which may be the same or different, are each hydrogen, optionally-substituted alkyl of 1 to 10 carbon atoms, or optionally-substituted phenyl, or R¹ is alkylthio of 1 to 6 carbon atoms; R is an ester-forming group: Y is oxygen or sulphur; and the carbamate grouping is in the 3- or 4-position on the phenyl ring), processes for their preparation and compositions containing them.

Acaricides

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TITLE MODIFIED
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This invention concerns acaricidal thiadiazoles, processes for their preparation, and compositions containing them.

5 In one aspect, the invention provides the thiadiazoles of the formula:



where: R^1 and R^2 , which may be the same or different, are each hydrogen, optionally-substituted alkyl of 1 to 10 carbon atoms, or optionally-substituted phenyl, or R^1 is alkylthio of 1 to 6 carbon atoms; R is an ester-forming group; Y is oxygen or sulphur; and the carbamate grouping is in the 3- or 4-position on the phenyl ring.

When R^1 or R^2 represents alkyl, it is preferably 20 of 1 to 6, especially of 1 to 4, carbon atoms, for example methyl, ethyl, n-propyl, isopropyl, n-butyl or t-butyl, and is desirably unsubstituted.

R^1 is preferably alkyl or alkylthio of 1 to 3 carbon atoms, or phenyl, specifically preferred groups being 25 ethyl, n-propyl, isopropyl, ethylthio and phenyl.

R^2 is preferably hydrogen.

The ester-forming group which R represents may be any suitable such group, but is preferably a substituted or unsubstituted alkyl group of 1 to 8 carbon atoms, a substituted or unsubstituted alkenyl or alkynyl group of 2 to 6 carbon atoms, a cycloalkyl group of 3 to 6 carbon atoms, or a substituted or unsubstituted aryl group. It is particularly preferred that R represents phenyl, an alkyl group, desirably of 1 to 4 carbon atoms, which is preferably unsubstituted, or a cycloalkyl group. When the group R represents an alkenyl group, it is preferably vinyl or allyl, and when it represents an alkynyl group it is preferably 1,1-dimethylpropargyl.

Particularly preferred groups which R may represent include methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, t-butyl, cyclopropyl, 1-methylheptyl, phenyl, α -methylbenzyl, and α -isopropylbenzyl.

When any of the groups R, R^1 and R^2 is substituted, it may be substituted for example by one or more halogen atoms, eg chlorine or bromine, or alkoxy groups of 1 to 4 carbon atoms.

Y is preferably oxygen.

The carbamate group is preferably attached to the phenyl ring in the 3-position.

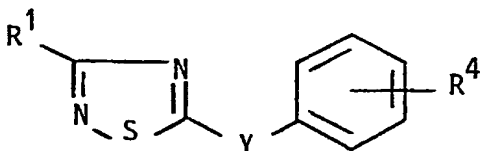
In a particularly preferred group of compounds

according to the invention, R^1 is ethyl, n-propyl, isopropyl, ethylthio or phenyl, R^2 is hydrogen, Y is oxygen, R is methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, t-butyl, cyclopropyl, 1-methylheptyl, phenyl, α -methylbenzyl or α -isopropylbenzyl, and the carbamate group is attached to the phenyl ring in the 3-position.

Specific preferred compounds of the invention are those of the Examples provided hereinafter. Particular mention may be made however of the compound [3-(3-n-propyl-1,2,4-thiadiazol-5-yloxy)phenyl]carbamate.

In another aspect, the invention provides a process for the preparation of a thiadiazole of formula I, in which a compound of the formula:

15



II

where R^1 and Y are as defined hereinbefore and R^4 represents $-NCO$ or $-NR^2.COCl$ and is in the 3- or 4-position on the phenyl ring, is reacted with an alcohol of the formula ROH where R is as defined hereinbefore, to give the desired compound.

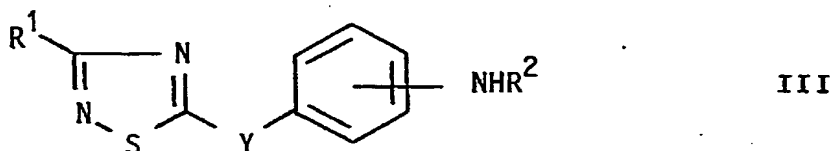
The reaction is conveniently effected by heating the

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isocyanate to a temperature of from 30 to 150°C with a molar excess of the alcohol, preferably under reflux.

The compounds of formula II may themselves be prepared by reacting the corresponding amines of the formula:

5



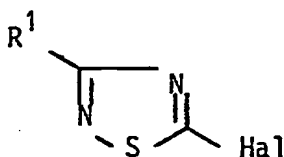
10 where R^1 , R^2 and Y are as defined hereinbefore, with phosgene in a suitable solvent medium to give the desired compound.

15 The reaction is preferably effected in an aromatic solvent which is a good solvent for the phosgene but does not react therewith, for example an aromatic hydrocarbon, eg toluene. The phosgene is desirably dissolved in the solvent prior to reaction with the amine, which is conveniently effected with heating to a temperature of from 30 to 150°C and preferably under reflux.

20 Where R^2 in the compound of formula III is hydrogen, the compound of formula II produced is one in which R^4 represents -NCO. Where R^2 in the compound of formula III is other than hydrogen, the compound of formula II produced is one in which R^4 represents $-NR^2.COCl$.

25 In turn, the amines of formula III may be prepared by

reaction of a thiadiazolyl halide of the formula:

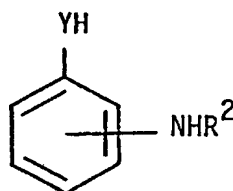


IV

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where R¹ is as defined hereinbefore, and Hal represents halogen, is reacted in the presence of a base with a phenylamine of the formula:

10



V

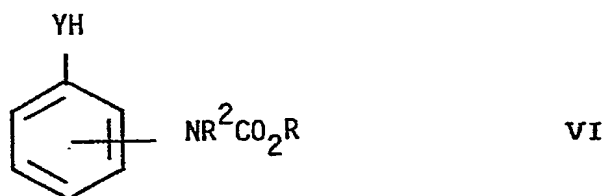
15 where R² and Y are as defined hereinbefore and the amino group is in the 3- or 4-position on the phenyl ring to give the desired compound.

The base employed is preferably a strong base, e.g. sodium hydride, and the reaction is conveniently effected
20 in a suitable inert solvent medium, for example dimethylformamide or tetrahydrofuran.

The thiadiazoles of formula I in which R² is other than hydrogen may be prepared by conventional alkylation processes from the corresponding compounds where R² is
25 hydrogen.

The compounds of formula I may alternatively be prepared by reaction in the presence of a base of a phenylcarbamate of the formula:

5



where R, R² and Y are as defined hereinbefore, and the carbamate group is in the 3- or 4- position on the phenyl ring, with a thiadiazolyl halide of formula IV as defined hereinbefore.

The base employed is preferably a strong base, e.g. sodium hydride, and the reaction is conveniently effected in a suitable inert solvent medium, for example dimethylformamide or tetrahydrofuran.

The compounds of formula VI may themselves be prepared by reaction of the appropriate aminophenol with a chloroformate of the formula ClCO₂R by methods known per se.

The thiadiazoles of formula I are of particular use against the eggs and larvae of acarids, particularly the eggs of the red spider mite, Tetranychus cinnabarinus, but also against the eggs and larvae of other mite species, e.g. Tetranychus urticae, Panonychus ulmi, Phyllocoptruta

oleivora, Eutetranychus banksi, Panonychus citri and
Tetranychus Mcdanieli.

Many of the compounds of the invention possess vapour
phase activity, which is a particular benefit where direct
5 contact with the acarids, their eggs or their larvae is
difficult to achieve.

The compounds of the invention are normally employed
in the form of compositions, which normally contain from
0.5 to 99%, preferably from 0.5 to 85% by weight, more
10 usually from 10 to 50% by weight, of the active compounds.
These are diluted if necessary before application to the
locus to be treated such that the concentration of active
ingredient in the formulation applied is from 0.05 to 5%
by weight.

15 The thiadiazoles of formula I are generally water
insoluble and may be formulated in any of the ways
commonly adopted for insoluble compounds.

For example, they may be dissolved in a water
immiscible solvent, for example a high boiling
20 hydrocarbon, as carrier, suitably containing dissolved
emulsifying agents so that the composition acts as a
self-emulsifiable oil on addition to water.

The thiadiazoles may alternatively be admixed with a
wetting agent with or without a solid carrier to form a
25 wettable powder which is soluble or dispersible in water.

or may be mixed with just a solid carrier to form a solid product.

An aqueous suspension concentrate may alternatively be prepared by grinding the compounds with water, a wetting agent and a suspending agent.

Solid carriers with which the substituted thiadiazoles may be incorporated include clays, sands, talc, mica or solid fertilizers, such products either comprising dust or larger particle size materials.

The surface active agents used may comprise anionic surface active agents, for example mono- or di-esters of phosphoric acid with fatty alcohol ethoxylates or salts of such esters, fatty alcohol sulphates, ethoxylated alkylphenol sulphates, lignin sulphonates, petroleum sulphonates, alkylaryl sulphonates such as alkyl-benzene sulphonates or lower alkyl-naphthalene sulphonates, salts of sulphonated naphthalene-formaldehyde condensates, salts of sulphonated phenol-formaldehyde condensates, or more complex sulphonates such as the amide sulphonates, e.g. the sulphonated condensation product of oleic acid and N-methyl taurine or the dialkyl sulphosuccinates e.g. the sodium sulphonates of dioctyl succinate.

The surface active agents may also comprise non-ionic agents, for example condensation products of fatty acid esters, fatty alcohols, fatty acid amides or

alkyl-substituted phenols with ethylene oxide, fatty esters of polyhydric alcohol ethers e.g. sorbitan fatty acid esters, condensation products of such esters with ethylene oxide, e.g. polyoxyethylene sorbitan fatty acid
5 esters, block copolymers of ethylene oxide and propylene oxide, acetylenic glycols such as 2,4,7,9-tetramethyl-5-decyn-4,7-diol, or ethoxylated acetylenic glycols.

The surface active agents may also comprise cationic agents, for example alkyl- and/or aryl-substituted
10 quaternary ammonium compounds such as cetyl trimethylammonium bromide, or ethoxylated tertiary fatty amines.

Preferred surface active agents include ethoxylated fatty alcohol sulphates, lignin sulphonates, alkyl-aryl
15 sulphonates, salts of sulphonated naphthalene-formaldehyde condensates, sodium oleoyl N-methyltauride, dialkyl sulphosuccinates, alkyl phenol ethoxylates, and fatty alkyl ethoxylates.

The composition may alternatively be in the form of an
20 aerosol composition, suitably using a cosolvent and a wetting agent, in addition to the propellant, which is suitably a polyhalogenated alkane such as dichloro-difluoromethane.

The compositions according to the present invention
25 may contain in addition to the substituted thiadiazoles

other active insecticides, acaricides, ovicides, bactericides and fungicides. It has been found that particular advantages are obtained with mixtures with other acaricides, e.g. clofentezine, but especially those
5 which combat the adult stages, e.g. amitraz, dicofol, cyhexatin or propargite, and particularly where the compound of formula I is one of those exemplified herein, especially isopropyl [3-(3-n-propyl-1,2,4-thiadiazol-5-yloxy)phenyl]carbamate.

10 The method of combating acarids, their eggs or their larvae provided by the present invention may be employed at any site where infestations of such pests are present or are liable or occur. Thus, it may be employed for example on plants or the soil.

15 Plants which may be treated include cotton and food crops, for example fruit trees and cereals, e.g. apples, pears, apricots, citrus fruits, maize, wheat, barley, beans, sugar beet, potatoes, carrots, or greenhouse crops, e.g. peppers, tomatoes, cucumbers, melons or strawberries.

20 In their various applications the compounds of formula I may be used at various rates; thus for example for the treatment of plants for the control of pests on plants the compounds are suitably applied at a rate of about 10-1000g per hectare or at a concentration of 1 to 2000ppm as
25 appropriate, e.g. 100 to 1000ppm, and preferably 50-300g

per hectare. Normally the compounds will be applied to the foliage of plants, but systemic activity may also be observed when applied to the soil around the base of the plants.

The following Examples are given merely to illustrate the present invention. The temperatures given therein are in °C, and parts and percentages are by weight.

Example 1

Isopropyl [3-(3-n-propyl-1,2,4-thiadiazol-5-yloxy)phenyl]-
10 carbamate

(a) 3-n-Propyl-5-(3-aminophenoxy)-1,2,4-thiadiazole

A solution of 32.7g of 3-aminophenol in 700ml dimethylformamide was stirred and treated slowly with 9g sodium hydride (80%), and was then heated to 50°C. A solution of
15 48.9g of 3-n-propyl-5-chloro-1,2,4-thiadiazole in 150ml dimethylformamide was then added dropwise, and the mixture was heated to 80°C with stirring for 5 hours. 3 Litres of water were added and stirred for ½ hour, following which the product was extracted into diethyl ether
20 (approximately 1 litre for 4 washings). After drying overnight over magnesium sulphate, the solution was filtered and evaporated, and 5% aqueous hydrochloric acid was added to form the hydrochloride salt. The aqueous solution of the salt was then extracted with diethyl
25 ether, and sodium bicarbonate was added to the aqueous

phase to pH 8. After further extraction into diethyl ether, and chromatography, the desired product was obtained as an oil.

(b) 3-n-Propyl-5-(3-isocyanatophenoxy)-1,2,4-thiadiazole

5 Phosgene was passed for 1½ hours into 650ml of toluene to form a saturated solution. To this was added 15.7g of the product of stage (a), and the mixture was stirred and heated to reflux for 1 hour. The solvent was then evaporated to yield 13.4g of the desired product.

10 (c) Isopropyl [3-(3-n-propyl-1,2,4-thiadiazol-5-yloxy)phenyl]carbamate

The product of stage (b) (4.17g) was refluxed for 2½ hours with 400ml of isopropanol. On evaporation of the solvent and recrystallisation from petroleum ether, the
15 desired product was obtained in a yield of 57%, mp. 64°C.

Examples 2-12

The following compounds of formula I where R² is hydrogen, Y is oxygen and the carbamate group is in the 3-position on the phenyl ring were prepared by methods
20 analogous to those of Example 1

<u>No.</u>	<u>R¹</u>	<u>R</u>	<u>M.Pt.</u>
2	methyl	methyl	117-119
3	methyl	isopropyl	69-71
4	ethyl	isopropyl	87-89
25	5 ethyl	α-isopropylbenzyl	93-95

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	<u>No.</u>	<u>R₁</u>	<u>R</u>	<u>M.Pt.</u>
	6	n-propyl	ethyl	52-53
	7	n-propyl	n-propyl	56
	8	n-propyl	n-butyl	43
5	9	n-propyl	isobutyl	92-94
	10	n-propyl	t-butyl	105-106
	11	n-propyl	phenyl	115
	12	n-propyl	α -isopropylbenzyl	99-101
	13	isopropyl	isopropyl	74-76
10	14	isopropyl	α -isopropylbenzyl	87-89
	15	phenyl	isopropyl	105
	16	phenyl	t-butyl	oil
	17	n-propyl	1-methylheptyl	oil
	18	n-propyl	α -methylbenzyl	oil
15	19	n-propyl	sec-butyl	86-88
	20	isopropyl	1,1-dimethylpropargyl	90-92

Examples 21-24

The following compounds of formula I where R² is hydrogen, Y is oxygen, and the carbamate group is in the 4-position on the phenyl ring, were prepared by methods analogous to that of Example 1:

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	<u>No.</u>	<u>R1</u>	<u>R</u>	<u>M.Pt.</u>
	21	n-propyl	isopropyl	73-74
	22	isopropyl	isopropyl	64-66
	23	ethylthio	isopropyl	117-119
5	24	ethylthio	t-butyl	97-99

Example 25

The following compound of formula I where R^2 is hydrogen, Y is sulphur, and the carbamate group is in the 3-position on the phenyl ring, was prepared by a method analogous to that of Example 1:

<u>No.</u>	<u>R1</u>	<u>R</u>	<u>M.Pt.</u>
25	n-propyl	isopropyl	oil

Example 26

Isopropyl [3-(3-n-propyl-1,2,4-thiadiazol-5-yloxy)phenyl]
 carbamate (alternative procedure)

(a) Isopropyl 3-hydroxyphenylcarbamate

A mixture of 3-aminophenol (109g), magnesium oxide (25g), water (350ml) and ethyl acetate (100ml) was stirred vigorously at room temperature while isopropyl chloroformate (123g) was added slowly. An ice bath was employed to ensure that the temperature did not exceed 30°C. After stirring for one hour at room temperature, the mixture was filtered to remove insoluble particles. These were washed thoroughly with ether, then the filtrates were combined, then separated, and the organic

- 15 -

layers were dried over magnesium sulphate. Evaporation under reduced pressure gave a viscous, pale yellow oil which was crystallised from toluene to give 162.7g of desired product, mp 75-77°C.

5 (b) Isopropyl [3-(3-n-propyl-1,2,4-thiadiazol-5-yloxy) phenyl]carbamate

A solution of the product of stage (a) (13.0g) and 3-n-propyl-5-chloro-1,2,4-thiadiazole (10.2g) in tetrahydrofuran (350ml), dried over a molecular sieve, was treated
10 with sodium hydride (80%, 2.0g), and was stirred and refluxed for 2 hours. It was then cooled to room temperature and was poured carefully into water (1 litre). After half an hour the ether extracts were separated, washed with water, and dried over sodium sulphate. The
15 solvent was removed to give a pale yellow oil which was recrystallised from 60-80° petrol to yield 14.2g of desired product as a white solid, mp 63-66°.

Example 27

A 40% suspension concentrate formulation of the
20 compound of Example 1 was prepared by blending the following ingredients:

25

- 15 -

	% w/w
Compound of Example 1	40
Polyfon H (sodium salt of sulphonated Kraft lignin - Westvaco Corp.)	2
5 Pluronic P75 (Ethylene oxide/propylene oxide block copolymer)	3
Xanthan Gum	0.2
Formaldehyde	0.2
Water	q.s.
10 Analogous 40% formulations were also prepared containing each of the compounds of Examples 2 to 25, as were analogous formulations respectively containing 0.5, 5, 10, 20, 50 and 85% by weight of the compound of Example 1 with corresponding changes in the amount of water	
15 employed.	

Example A

Discs (2cm diameter) were cut from the lamina of the first true leaves of dwarf French bean plants, Phaseolus vulgaris. The discs were placed on moist 3cm wide strips of filter paper, and 10-15 adult female mites (Tetranychus cinnabarinus) were placed on each disc. These were allowed to lay eggs for a period of 24 hours, and were then removed. The discs were then dipped into solutions of the compounds of the Examples listed below at various concentrations prepared by dissolving the appropriate

quantity of the compound in 2ml acetone plus 5% Atlox 1025A wetting agent and diluting the solution with water containing Synperonic N wetting agent so as to give an aqueous formulation containing 0.5ml/litre Synperonic N, 5 20ml/litre acetone and 1ml/litre Atlox 1025A.

Two replicates were used per treatment and the number of eggs hatching within 8 days was assessed.

The results below show the lowest rate tested at which the compounds enumerated killed at least 75% of the eggs.

	<u>Compound Ex. No.</u>	<u>Rate (ppm)</u>
10	1	1
	4	64
	6	26
	7	10
15	8	1.6
	9	10
	10	0.64
	11	1.6
	12	1.6
20	13	4
	14	26
	16	160
	17	4
	18	26
25	19	4

<u>Compound Ex. No.</u>	<u>Rate (ppm)</u>
20	64
21	160
23	64

5

10

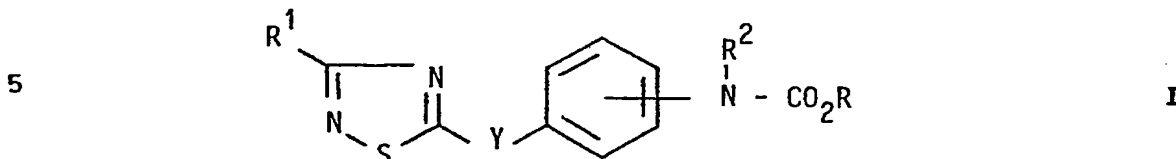
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CLAIMS

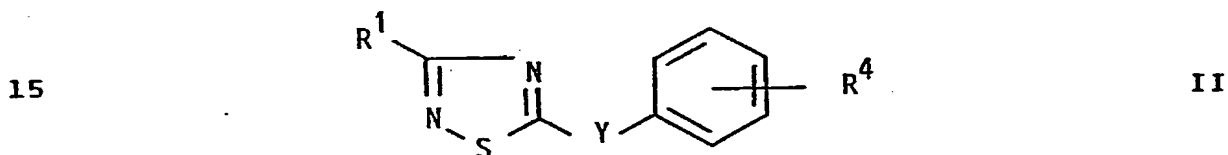
1. The thiadiazoles of the formula:



- where: R^1 and R^2 , which may be the same or different, are each hydrogen, optionally-substituted alkyl of 1 to 10 carbon atoms, or optionally-substituted phenyl, or R^1 is alkylthio of 1 to 6 carbon atoms; R is an ester-forming group; Y is oxygen or sulphur; and the carbamate grouping is in the 3- or 4-position on the phenyl ring.
- 10
- 15 2. The thiadiazoles according to claim 1 wherein R^1 is phenyl or alkyl of 1 to 6 carbon atoms.
3. The thiadiazoles according to claim 1 or claim 2 wherein R is a substituted or unsubstituted alkyl group of 1 to 8 carbon atoms, a substituted or unsubstituted alkenyl or alkynyl group of 2 to 6 carbon atoms, a cycloalkyl group of 3 to 6 carbon atoms, or a substituted or unsubstituted aryl group.
- 20
4. The thiadiazoles according to any of claims 1 to 3 wherein the carbamate group is attached to the phenyl ring in the 3-position.
- 25

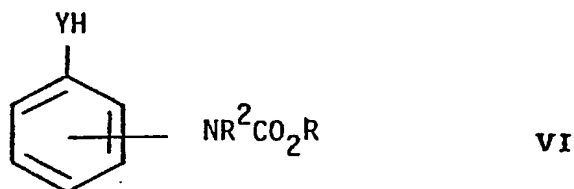
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5. The thiadiazoles according to any of claims 1 to 4 wherein R^1 is ethyl, n-propyl, isopropyl, ethylthio or phenyl, R^2 is hydrogen, Y is oxygen, R is methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, t-butyl, cyclopropyl, 1-methylheptyl, phenyl, α -methylbenzyl or α -isopropylbenzyl, and the carbamate group is attached to the phenyl ring in the 3-position.
6. [3-(3-n-Propyl-1,2,4-thiadiazol-5-yloxy)phenyl]carbamate.
7. A process for the preparation of a thiadiazole of formula I according to claim 1, in which a compound of the formula:



- where R^1 and Y are as defined in claim 1 and R^4 represents $-NCO$ or $-NR^2.COCl$ and is in the 3- or 4-position on the phenyl ring, is reacted with an alcohol of the formula ROH where R is as defined in claim 1, to give the desired compound.
8. A process for the preparation of a thiadiazole of formula I according to claim 1, in which a phenylcarbamate of the formula:

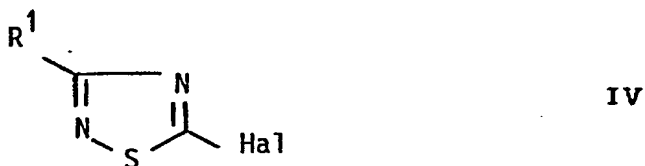
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5

where R , R^2 and Y are as defined in claim 1, and the carbamate group is in the 3- or 4- position on the phenyl ring, is reacted in the presence of a base with a thiadiazolyl halide of the formula:

10



15

where R^1 is as defined in claim 1 and Hal represents halogen, to give the desired compound.

9. An acaricidal composition which comprises from 0.5 to 99% by weight of one or more thiadiazoles according to claim 1 in association with a suitable carrier and/or surface active agent.

20

10. A method of combating acarids, their eggs or their larvae which comprises applying to a locus infested or liable to be infested therewith an effective amount of one or more thiadiazoles according to claim 1.

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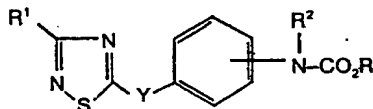
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EP 0 200 334 A3



European Patent
Office

EUROPEAN SEARCH REPORT

0200334

Application number

EP 86 30 2066

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
E	EP-A-O 177 710 (BAYER) * Whole document; page 3, formule V; pages 15,21,22,40 * -----	1-8	C 07 D 285/08 A 01 N 47/20
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			C 07 D 285/00
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 09-07-1987	Examiner ALLARD M. S.
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	